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Consistency of Urban Background Black Carbon Concentration Measurements by Portable AE51 and Reference AE22 Aethalometers: Effect of Corrections for Filter Loading

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ABSTRACT

Monitoring exposure to black carbon with portable devices is an important part of researching the health impacts of combustion-related air pollutants. We collected 786 hourly averaged equivalent black carbon (eBC) measurements from co-located duplicate portable AE51 Aethalometers and a UK Government reference AE22 Aethalometer (the data for the latter were corrected for filter darkening effects using a standard procedure), at an urban background site in Glasgow, UK. The AE51 and the reference concentrations were highly correlated ($R^2 \geq 0.87$) for the combined deployment periods. The application of a previously reported method for correcting the AE51's underestimation of concentrations, associated with filter loading, generally led to an overestimation of values (specifically, the normalised mean bias values for the two AE51s increased from -2% and $+3\%$ to $+14\%$ and $+25\%$ across the full range of measurements after correction). We found only limited and inconsistent evidence that the AE51 Aethalometers (attenuation [AE51_ATN] $\leq \sim 52$) underestimated the eBC concentrations compared to the reference measurements. Thus, our observations indicate that the AE51 can achieve close agreement with the reference AE22 monitor without applying corrections for filter loading at relatively low AE51_ATN values in environments with low eBC concentrations.

Keywords: Air pollution; Equivalent black carbon; Aethalometer; Micro-Aethalometer; Attenuation; Filter loading.

INTRODUCTION

Black carbon (BC) is a constituent of airborne particulate matter (PM) produced during incomplete combustion of carbon-based fuels. The health effects associated with exposure to PM and BC include respiratory and cardiovascular diseases (WHO, 2005, 2013), including associations between health outcomes and proximity to roads where BC concentrations are frequently elevated (Janssen *et al.*, 2011, 2012; Grahame *et al.*, 2014). In the UK, BC concentrations are measured continuously at 14 sites in a nationally coordinated network (<https://uk-air.defra.gov.uk/networks/network-info?view=ukbsn>) using a mains-powered rack-mounted Aethalometer (AE22; Magee

Scientific, CA, USA). The number of fixed-site BC measurements is limited because of the high installation, equipment and maintenance costs and because BC does not need to be measured under current UK air quality compliance legislation.

Battery-powered, hand-held Aethalometers have also been developed to measure real-time concentrations of BC. The small size and light weight of these monitors make them suitable for use in a variety of applications including static locations in a network (Weichenthal *et al.*, 2014; Montagne *et al.*, 2015; Gillespie *et al.*, 2016), mobile monitoring (Apte *et al.*, 2011; Hankey and Marshall, 2015; Van den Bossche *et al.*, 2015) and/or personal monitoring (Dons *et al.*, 2012, 2013a, b; Williams and Knibbs, 2016). In this study, we evaluated two microAeth AE51 portable BC Aethalometers (AethLabs; San Francisco, CA, USA).

Following recommendations made by Petzold *et al.* (2013), in the remainder of this paper we use the term equivalent black carbon (eBC) to describe BC concentrations quantified by the optical absorption technique used in Aethalometer

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instruments. In this technique air is sampled through a filter and the concentration of BC is estimated by comparing the attenuation (ATN) of light passing through the particles deposited on the filter to that passing through an unloaded reference point on the same filter. An important consideration is that the relationship between ATN and BC loading is not linear at higher attenuation values. Different methods to correct Aethalometer data to account for these filter loading effects have been proposed (Kirchstetter and Novakov, 2007; Virkkula *et al.*, 2007) but few studies have compared the correction algorithms to determine if they are always necessary, and which provides the most accurate correction (Good *et al.*, 2017).

The aim of our study was to compare methods commonly used to correct BC measurements from AE51 Aethalometers and to establish how consistent the measurements from these portable systems are with a static Aethalometer used in the UK Government black carbon network. We build on the study of Good *et al.* (2017), who evaluated correction algorithms for filter darkening, by evaluating some of these correction methods during repeated AE51 Aethalometer field deployments. Good *et al.* (2017) used an online Photoacoustic Extinctionmeter (PAX; Droplet Measurement Technologies, Boulder, CO, USA) as a reference instrument under controlled laboratory conditions. In contrast, we deployed duplicate AE51 Aethalometers close to the inlet of an AE22 ‘reference’ Aethalometer at an urban background site in the city of Glasgow, UK, for 786 hours of co-located measurements interspersed between April and August 2016. The static outdoor deployment of the AE51 Aethalometers avoided the potential introduction of large spurious readings due to mechanical shocks that has been observed during mobile monitoring (Apte *et al.*, 2011). The static deployment also minimised another potential source of error sometimes experienced during personal monitoring: the condensation of water on the AE51 monitor filters or optics arising from rapid changes in temperature and/or relative humidity (Cai *et al.*, 2013). Since these two source of potential error were reduced in our study, we were able to focus our field-based assessment of filter loading effects directly on the agreement between measurements from portable and reference Aethalometer instruments.

METHODS

Site Description; AE22 Aethalometer Operation

Measurements were made at the Glasgow Townhead monitoring site, an urban background location in central Glasgow (55.866°N, 4.244°W) that is part of the UK black carbon monitoring network. Hourly averaged reference eBC measurements made at this site using a Magee Scientific AE22 Aethalometer are publicly available (https://uk-air.defra.gov.uk/data/data_selector). AE22 instrument operation and data ratification are subject to national QA/QC protocols (Butterfield *et al.*, 2015). Prior to publication, the AE22 hourly averaged reference concentration data are corrected for filter darkening effects estimated from changes in quantified concentrations at the time of automatic filter advances using a standard

correction procedure (Virkkula *et al.*, 2000; Butterfield *et al.*, 2015).

AE51 Aethalometer Set-up and Operation

Two microAeth AE51 Aethalometers (<https://aethlabs.com/>), subsequently referred to as ‘BC_1204’ and ‘BC_1303’, were deployed in waterproof boxes on the roof of the monitoring station and sampled ambient air through a 1-m length of tubing supplied by the manufacturer. A conductive asbestos sampling inlet (SKC Ltd, UK) was connected to the inlet of the tubing as a rain hood to prevent water ingress. The flow rate of the AE51 Aethalometers was set to 50 mL min⁻¹ and data were recorded each minute.

The AE51 Aethalometers were co-located at the monitoring station on 8 occasions in 2016 (Table 1). During each deployment the site was visited approximately every 5 days to download the AE51 data and to change filters. The AE51 filter attenuation did not exceed 52.01 during any deployment period.

The 1-min data collected by the AE51 Aethalometers were averaged to hourly concentrations for comparison with reference AE22 eBC concentrations.

Correction of AE51 Equivalent Black Carbon Data

Prior to filter darkening correction procedures we smoothed the AE51 data to minimise the number of negative values using Optimized Noise-reduction Algorithm (ONA) software from the AethLabs website (<https://aethlabs.com/dashboard>) (Hagler *et al.*, 2011). We set the change in attenuation value used to average eBC concentration data in this ONA method to 0.05.

We then applied the correction procedure published by Apte *et al.* (2011) based on Kirchstetter and Novakov (2007) (correction subsequently referred to as *K&N* in our paper) to the ONA-adjusted eBC data from the portable AE51 Aethalometers to account for potential underestimation of eBC as the darkness of the filter increased:

$$eBC_{corrected} = eBC_{ONA}((0.88 \times \exp^{-ATN/100}) + 0.12)^{-1} \quad (1)$$

where eBC_{ONA} is the AE51 concentration after ONA correction (outlined above) and ATN is the attenuation of the AE51 filter. We used the ATN values directly from the AE51 monitors as clean filters were used on each occasion, in contrast to Good *et al.* (2017), who used the percent change in attenuation between the start and end of the study to account for the use of preloaded filters.

Virkkula *et al.* (2007) developed a correction procedure based on the concept that increasing ATN results in a linear underestimation of the correct eBC concentration:

$$eBC_{corrected} = (1 + k \times ATN)eBC_{ONA} \quad (2)$$

We estimated values of k from the linear regression slope between the ratio of AE22 Aethalometer concentrations divided by AE51 eBC_{ONA} concentrations vs. corresponding AE51 ATN values; with a fixed intercept value of 1 (Cheng and Lin, 2013).

Table 1. Dates and descriptive statistics for co-located AE51 and reference AE22 Aethalometer deployments at the Glasgow Townhead network monitoring site. Data are based on hourly averages. k values represent the slope of the ratio of AE22/AE51 vs. AE51_ATN as outlined in 'METHODS'.

Period	AE22				BC1024				BC1303			
	N	Min BC (AE22)	Max BC (AE22)		Min ATN (AE51)	Max ATN (AE51)	R^2	k	Min ATN (AE51)	Max ATN (AE51)	R^2	k
29/04–04/05/16	114	0.1	2.1		−1.12	24.07	0.90	0.0028	3.20	27.97	0.85	0.0054
04/05–09/05/16	117	0.1	3.9		−1.50	44.87	0.93	−0.0012	−0.54	48.64	0.92	−0.0027
09/05–13/05/16	94	0.2	1.7		2.64	37.93	0.83	0.0004	3.18	39.54	0.81	−0.0003
18/05–22/05/16	95	0.1	1.9		−6.17	20.28	0.94	0.0107	0.99	30.05	0.93	0.0032
23/05–26/05/16	68	0.1	1.2		−4.36	14.59	0.85	0.0157	0.58	19.11	0.80	0.0017
08/08–12/08/16	88	0	3.5		−3.08	21.92	0.98	0.0065	6.18	33.85	0.98	−0.0003
12/08–17/08/16	117	0.1	2.4		−1.07	37.64	0.95	0.0049	2.02	43.51	0.96	−0.00005
17/08–22/08/16	93	0.2	3.6		−0.33	43.97	0.96	0.0010	0.24	52.01	0.94	−0.0010
Mean k for above 8 periods:								0.0051				0.0007
Std dev k for above 8 periods:								0.0057				0.0026
Relative standard deviation k (%):								112				346
k for complete dataset:								0.0017				−0.000009

In an equivalent way as was described for uncorrected 1-min A51 data in the preceding section, corrected 1-min AE51 Aethalometer data were averaged to hourly concentrations for comparison with reference AE22 eBC concentrations.

RESULTS

Precision and Accuracy of AE51 Aethalometers

Unadjusted concentrations measured by the two duplicate AE51 Aethalometers tracked each other closely over extended time periods (regression analyses of AE51_1303 vs. AE51_1204: slope = 1.06, intercept = 0.00, $n = 786$, $R^2 = 0.97$) (Fig. 1) with both instruments recording similar overall average values (0.66 and 0.69 $\mu\text{g m}^{-3}$ for AE51_1204 and AE51_1303 respectively; Table 2). The mean absolute percentage error and normalised mean absolute error for duplicate measurements (Fig 1) were both 5% (following normalisation of observations from individual instruments by the average of duplicate measurements). Similarly high correlations between duplicate AE51 Aethalometers have been reported in other studies, e.g., R^2 values > 0.95 between 13 co-located monitors (Dons *et al.*, 2012).

Time series of hourly averaged unadjusted concentrations from the duplicate AE51 Aethalometers and the reference AE22 Aethalometer also showed very similar temporal patterns (Figs. 2(a)–2(d)) with R^2 values between AE51_1204 vs. AE22 and AE51_1303 vs. AE22 of 0.88 and 0.87 respectively (Figs. 2(e) and 2(f)). A previous study at a BC network monitoring site in Birmingham, UK (Delgado-Saborit, 2012), reported high correlation ($R^2 = 0.90$) between AE51 and reference AE22 Aethalometer concentrations over a similar range of reference analyser BC concentrations (0–5 $\mu\text{g m}^{-3}$) to the BC concentrations observed during our deployments.

Effect of Filter Loading Corrections

Examination of scatterplots of the difference between AE51 and AE22 concentrations vs. AE51_ATN provided no obvious indication of reduction in measured BC by AE51 instruments cf. AE22 instruments as the ATN of the former increased between 0–52 (Figs. 2(g)–2(h)). The correlation between AE51–AE22, and AE51_ATN, was very low ($R^2 < 1\%$ for both AE51 instruments), with no evidence of a slightly negative slope that might be expected from relative underestimation by AE51 as AE51_ATN increased. Almost identical findings were noted for hourly averaged ONA-adjusted AE51 observations (Fig. 3) consistent with the ONA adjustment mainly affecting short-term fluctuations that were removed by averaging over hourly periods.

In contrast ONA- and $K\&N$ -adjusted AE51 observations substantially overestimated AE22 observations (e.g., AE51 vs. AE22 regression lines of $1.04AE22 + 0.6$ and $1.16AE22 + 0.05$; Figs. 4(a)–4(f)), with clear indication that the $K\&N$ -adjusted AE51 overestimation increased with increasing AE51_ATN (Figs. 4(g)–4(h)).

We examined ratios of AE22/AE51 for possible underestimation of eBC by AE51 instruments as AE51_ATN

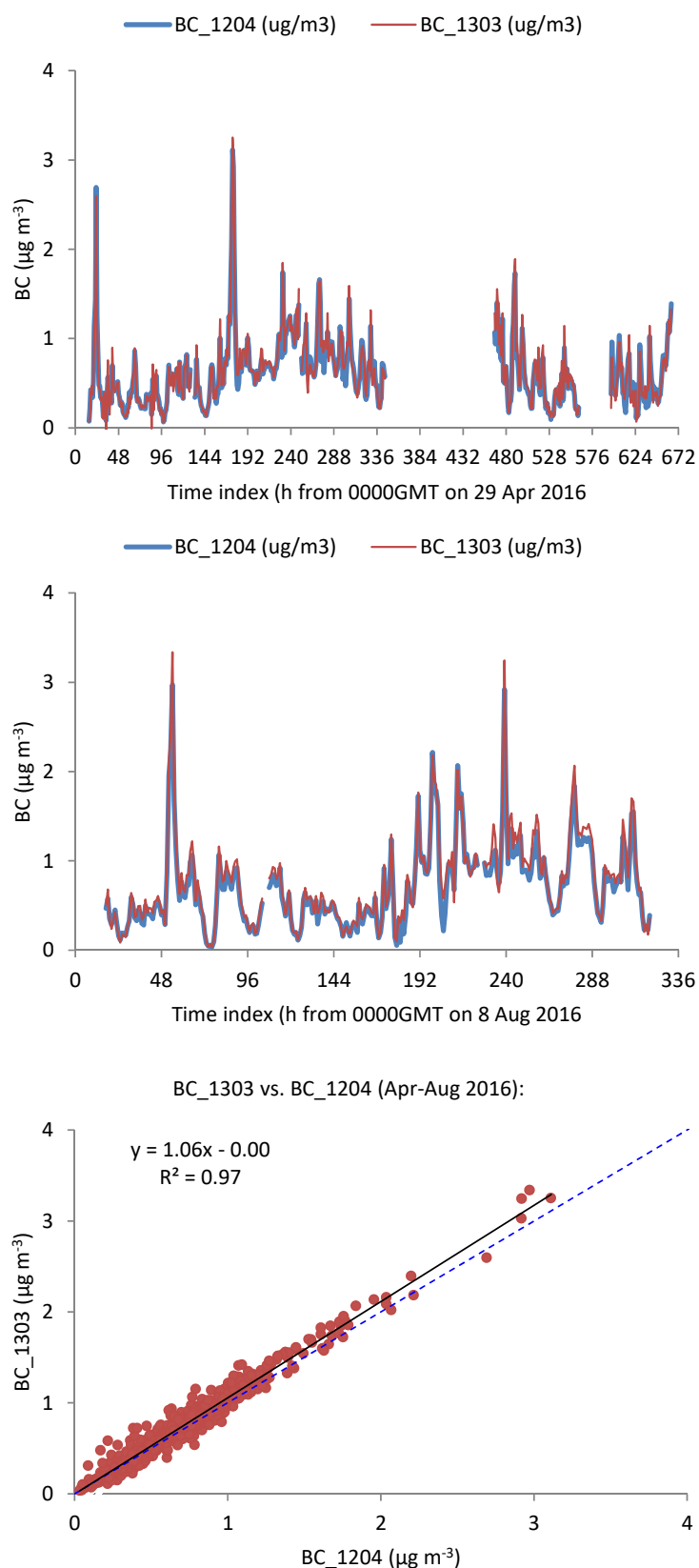


Fig. 1. (a) Time series of unadjusted hourly averaged eBC concentrations measured by two AE51 micro-Aethalometers (BC_1204 and BC_1303) between 29/04/16 and 26/05/16. (b) Time series of unadjusted hourly averaged eBC concentrations measured by BC_1204 and BC_1303 between 08/08/16 and 22/08/16. Gaps in plot separate non-consecutive time series. (c) Scatter plot with linear regression of hourly averaged unadjusted concentrations measured during both of the above periods by duplicate AE51 Aethalometers. The dashed line represents 1:1 correspondence.

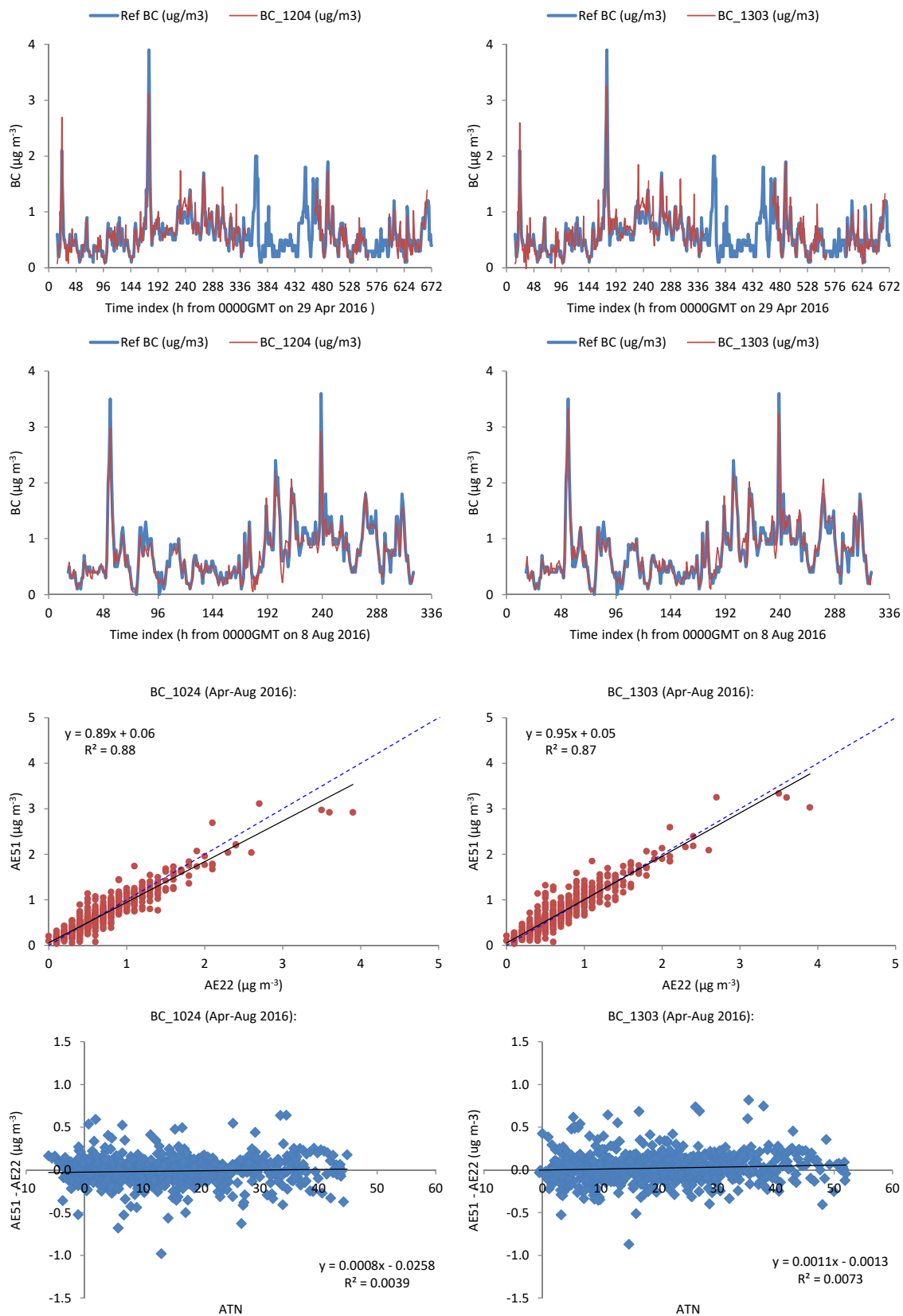


Fig. 2. (a–d) Time series of adjusted hourly averaged eBC concentrations from reference AE22 Aethalometer and unadjusted concentrations measured by two AE51 micro-Aethalometers for 29/04/16–26/05/16 and 08/08/16–22/08/16. (e–f) Scatter plots of unadjusted eBC concentrations from AE51 micro-Aethalometers vs. adjusted eBC concentrations measured by the AE22 Aethalometer. Dashed lines represent 1:1 correspondence. (g–h) Scatter plots of difference between AE51_eBC–AE22_eBC vs. AE51_ATN. Panels (a–h) in this figure are arranged from left to right, top to bottom.

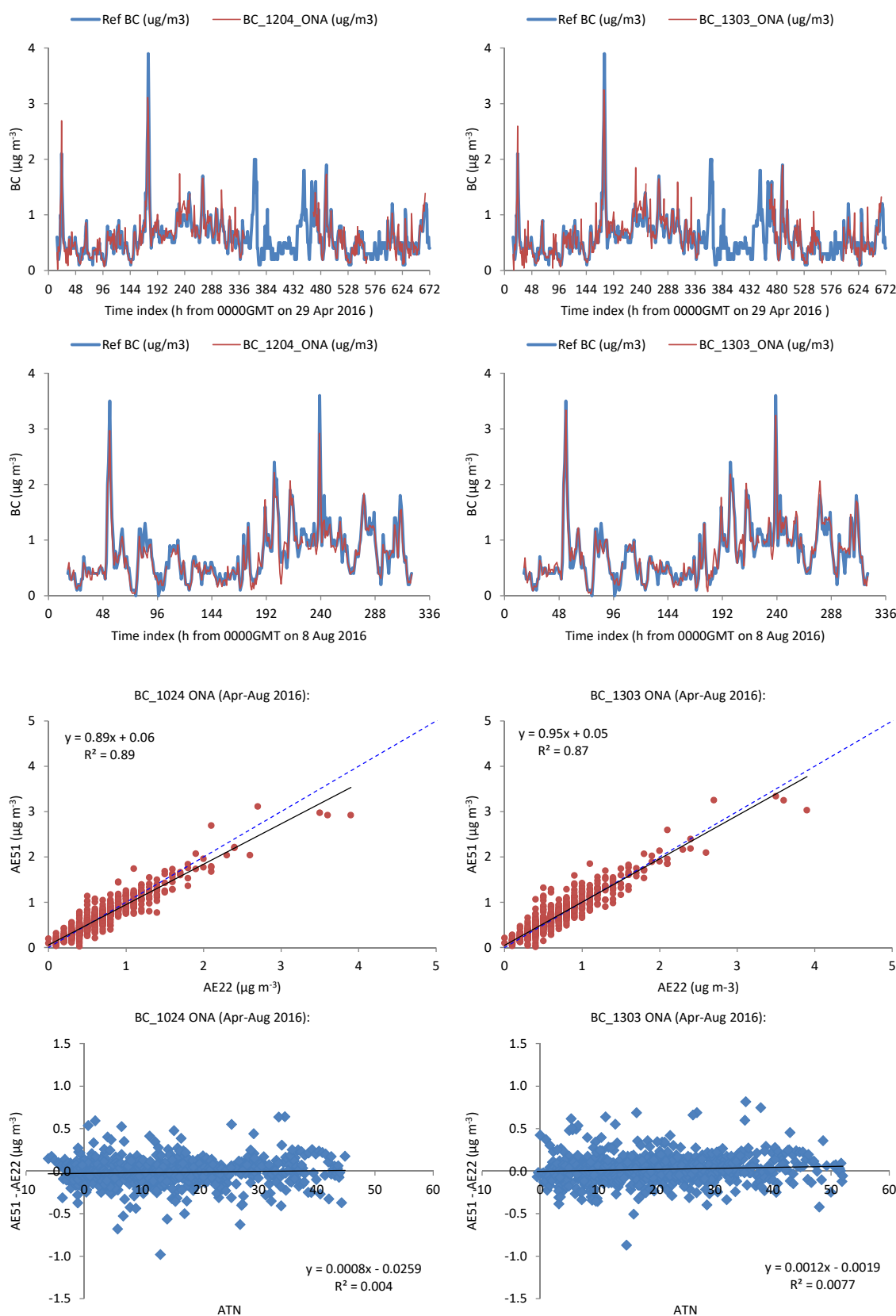


Fig. 3. (a–d) Time series of adjusted hourly averaged eBC concentrations from reference AE22 Aethalometer and ONA-adjusted concentrations from two AE51 micro-Aethalometers for 29/04/16–26/05/16 and 08/08/16–22/08/16. (e–f) Scatter plots of ONA-adjusted eBC concentrations from AE51 BC micro-Aethalometers vs. adjusted eBC concentrations measured by the AE22 Aethalometer. Dashed lines represent 1:1 correspondence. (g–h) Scatter plots of difference between ONA-adjusted AE51_BC–AE22_eBC vs. AE51_ATN. Panels (a–h) in this figure are arranged from left to right, top to bottom.

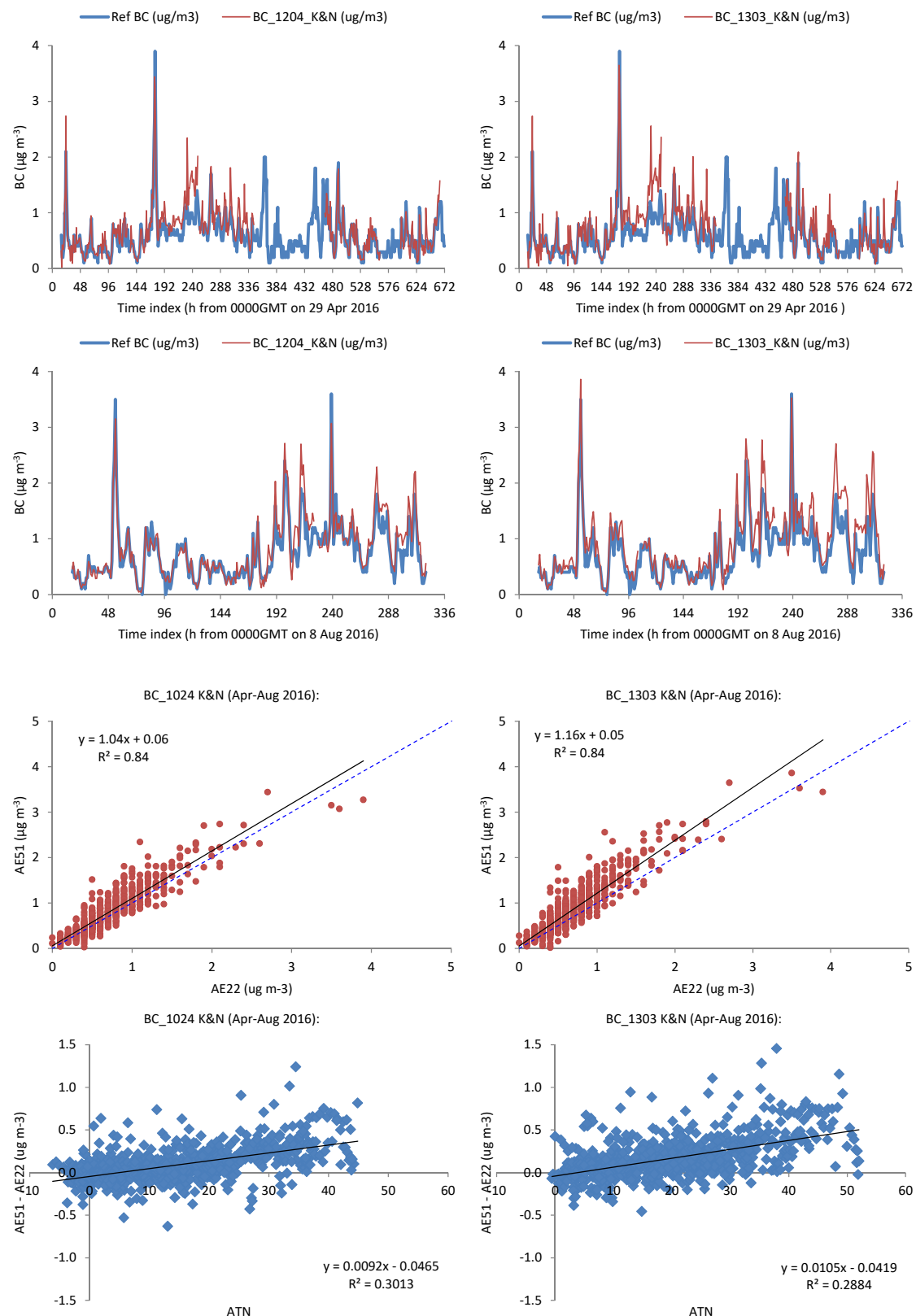


Fig. 4. (a–d) Time series of adjusted hourly averaged eBC from reference AE22 Aethalometer and consecutive ONA- and K&N-adjusted concentrations from two AE51 micro-Aethalometers for 29/04/16–26/05/16 and 08/08/16–22/08/16. (e–f) Scatter plots of consecutive ONA- and K&N-adjusted eBC concentrations from AE51 eBC vs. adjusted eBC concentrations measured by the AE22 Aethalometer. Dashed lines represent 1:1 correspondence. (g–h) Scatter plots of difference between ONA- and K&N-adjusted AE51_eBC–AE22_eBC vs. AE51_ATN. Panels (a–h) in this figure are arranged from left to right, top to bottom.

increased (Fig. 5). When all available data were included in the regression analyses of AE22/AE51 vs. AE51_ATN the values of k calculated were 0.0017 and -0.000009 for 1-h average data for BC_1204 and BC_1303 monitors respectively (Fig. 6, Table 1). k values calculated for the 8 individual measurement periods (Fig. S1) had overall averages (and relative standard deviations) of 0.0051 (RSD = 112%) and 0.0007 (RSD = 346%) (Table 1, Fig. S2). Negative k values suggest that non-corrected eBC concentrations overestimate the ‘true’ eBC concentrations and have been suggested to occur in summer months when the ratio of black carbon to aerosol volume concentrations is lower (Virkkula *et al.*, 2007). The range of k values that we calculated was not inconsistent with calculations in other studies, e.g., $k = 0.0039$ (Cheng and Lin, 2013), $k = 0.0033$ (Cheng *et al.*, 2014) and $k = 0.01$ (Morales Betancourt *et al.*, 2017).

We then corrected eBC_{ONA} values, using Eq. (2) with both: the average k values calculated for the 8 individual measurement periods, and the k values calculated for the complete dataset (i.e., all 8 periods grouped together). Data adjusted in this way using average and complete-dataset k values were only marginally different from the unadjusted

data with only minor changes in statistics comparing the adjusted data against reference AE22 observations—with the average k value cases resulting in minor deterioration in the closeness of fit between adjusted and observed data (Table 2).

The increased regression slopes for $K\&N$ -corrected AE51 data was reflected in prediction model statistics that illustrated the scale of overestimation (e.g., NMB statistics of 14% and 25%; Table 2). The $K\&N$ -corrected hourly AE51 Aethalometer concentrations had the largest NRMSE statistics (NRMSE = 34% and 44% for BC_1204 and BC_1303 respectively; Table 2). Hence the $K\&N$ equation overcorrected the AE51 Aethalometer concentrations for filter loading effects, with lower correlation against the reference Aethalometer concentrations than the unadjusted AE51 data (Table 1).

DISCUSSION

Portable AE51 Aethalometers are widely used in mobile and personal monitoring. However, field evaluations comparing the AE51 to ‘reference’ eBC monitoring instruments have been limited. In our study, we found that

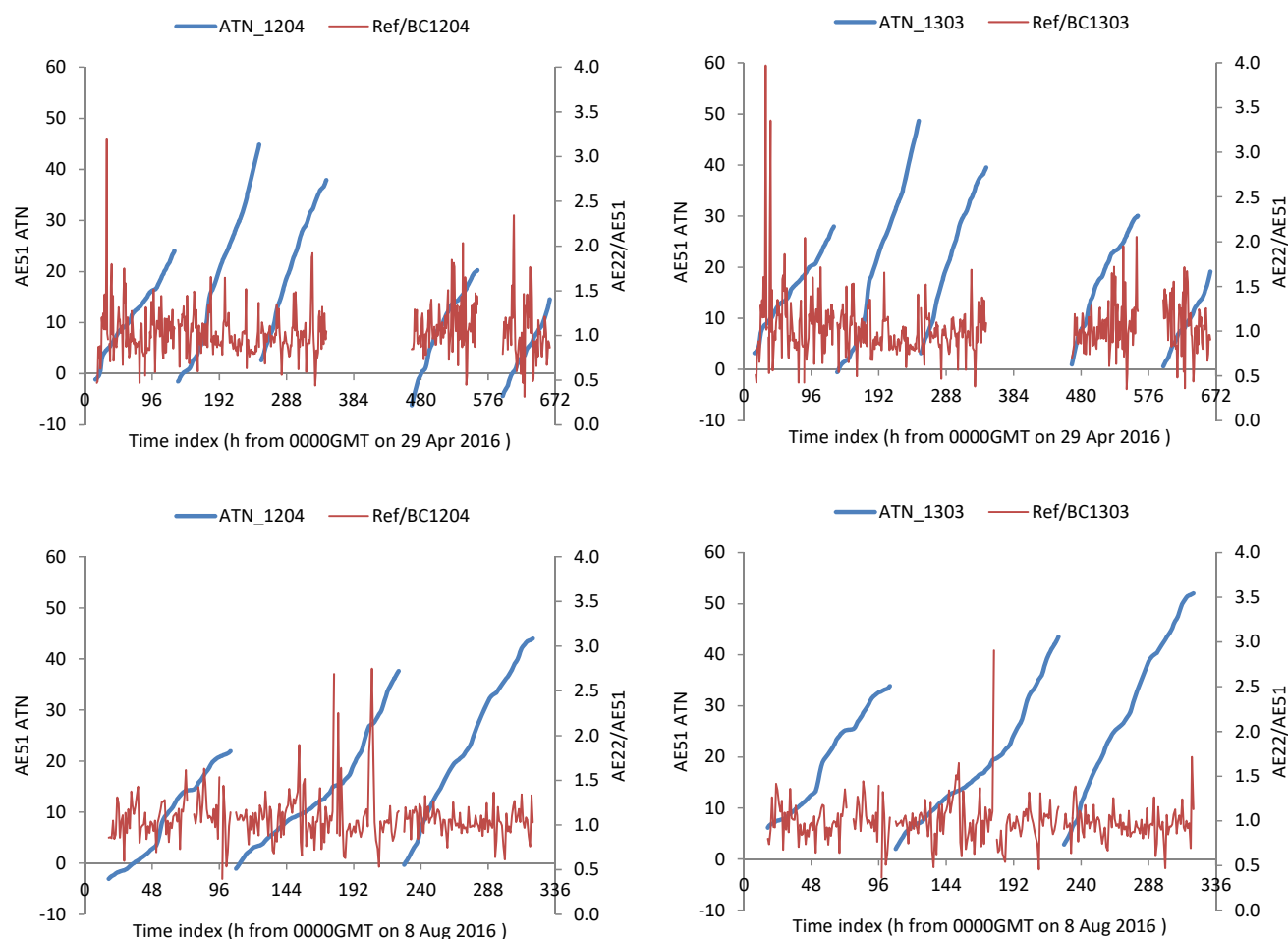


Fig. 5. Time series of ATN from two AE51 micro-Aethalometers and ratio of adjusted hourly averaged eBC concentrations from reference AE22 Aethalometer and AE51 micro-Aethalometers for (a and b) 29/04/16–26/05/16 and (c and d) 08/08/16–22/08/16.

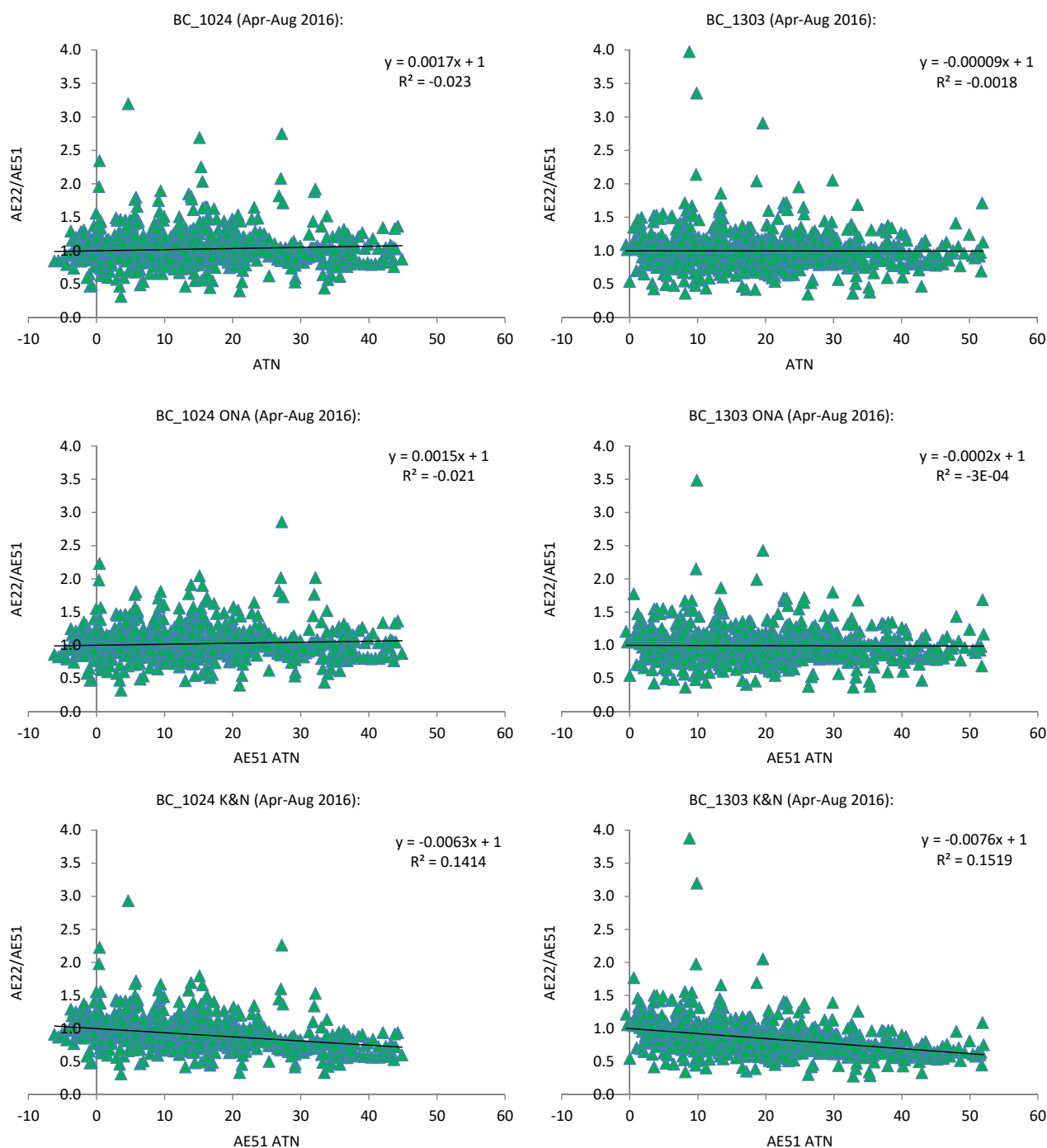


Fig. 6. Scatter plots of hourly averaged eBC concentration ratios for reference AE22 Aethalometer (adjusted)/AE51 micro-Aethalometers (non-adjusted and adjusted) for 29/04/16–26/05/16 and 08/08/16–22/08/16. Panels (a–h) in this figure are arranged from left to right, top to bottom.

the coefficients of determination for hourly averaged data between two AE51s and a reference AE22 Aethalometer, were 87% and 88%, which are similar to the R^2 values (> 0.75) reported for 10-min-averaged data between an AE51 and a Thermo Multi-Angle Absorption Photometer (MAAP) for a dataset with a minimum concentration of $1.5 \mu\text{g m}^{-3}$ (Viana *et al.*, 2015).

Applying the *K&N* correction for filter darkening to the AE51 data resulted in overestimated eBC values (based on

the reference data) and larger errors (compared to the unadjusted measurements) (Fig. 4, Table 1). In a previous chamber experiment comparing an AE51 and a Photoacoustic Extinctionmeter, the former underestimated concentrations when using unloaded filters (Good *et al.*, 2017); however, when pre-loaded filters were used, the *K&N* correction overcompensated for filter loading effects. The results from our study indicate that this correction leads to overestimated AE51 values (cf. reference AE22 concentrations) during

Table 2. Agreement between co-located AE51 and reference AE22 BC Aethalometer instruments at the Glasgow Townhead network monitoring site. Units of average (Ave), mean bias (MB), mean absolute error (MAE), root mean square error (RMSE) and intercept are $\mu\text{g m}^{-3}$. Normalised mean bias (NMB), normalised mean absolute error (NMAE), normalised root mean square error (NMRSE), slope and R^2 are dimensionless. Statistics were normalised by arithmetic mean of hourly reference AE22 instrument measurements ($0.67 \mu\text{g m}^{-3}$). *BC1024_Corr(ave)* and *BC1303_Corr(ave)* represent eBC corrected using average k values from 8 individual measurement periods. *BC1024_Corr(comp)* and *BC1303_Corr(comp)* represent eBC corrected using k values from complete dataset (i.e., all 8 measurement periods grouped together).

	Ave	MB	NMB	MAE	NMAE	RMSE	NMRMSE	Slope	Intercept	R^2
BC1024	0.66	−0.01	−0.02	0.10	0.16	0.15	0.23	0.89	0.06	0.88
BC1024 ONA	0.66	−0.01	−0.02	0.10	0.16	0.15	0.23	0.89	0.06	0.89
BC1024 <i>K&N</i>	0.76	0.09	0.14	0.15	0.23	0.22	0.34	1.04	0.06	0.84
BC1024_Corr(ave)	0.71	0.04	0.06	0.12	0.18	0.17	0.26	0.97	0.06	0.87
BC1024_Corr(comp)	0.67	0.00	0.01	0.10	0.16	0.15	0.23	0.92	0.06	0.87
BC1303	0.69	0.02	0.03	0.11	0.17	0.16	0.25	0.95	0.05	0.87
BC1303 ONA	0.69	0.02	0.03	0.11	0.17	0.16	0.25	0.95	0.05	0.87
BC1303 <i>K&N</i>	0.83	0.16	0.25	0.15	0.23	0.28	0.44	1.16	0.05	0.84
BC1303_Corr(ave)	0.70	0.03	0.05	0.11	0.18	0.18	0.28	0.97	0.05	0.87
BC1303_Corr(comp)	0.69	0.02	0.03	0.11	0.17	0.16	0.25	0.95	0.05	0.87
AE22:	Ave:	Min:	Max:							
	0.67	0.10	3.90							

conditions with relatively low concentrations and low filter loads (AE51_ATN < 52).

Our study has several important limitations, including being specific to a single geographical location during a relatively short time period. Also, we did not have access to the raw measurements from the reference instrument, as the data published on the governmental website, i.e., the reference concentrations, had already been corrected according to a standard procedure (by Virkkula *et al.*). Further research on the agreement between AE51 and AE22 eBC measurements, at locations where more detailed data may be available, could assess the influence of particle composition, meteorological conditions, and the numerical magnitude of filter loading corrections to AE22 data.

In summary, we compared field measurements of eBC concentrations at an urban background site obtained with two portable AE51 Aethalometers vs. a reference AE22 Aethalometer. The uncorrected AE51 measurements were in close agreement with the reference concentrations, with a slight overall underestimation by one AE51 (NMB = −2%) and a slight overall overestimation by the other (NMB = +3%). After correcting for filter loading, the data from both AE51 Aethalometers generally overestimated the reference concentrations (NMB = +14% and +25%). These results suggest that the AE51 Aethalometer measurements may not require correction for filter loading in order to maintain consistency with reference AE22 concentrations when the AE51_ATN values are less than 52.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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